

Natural peloids *versus* designed and engineered peloids

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Abstract

The present study pretends to show what distinguishes *natural peloids* from *artificial peloids*. The last *peloids* referred to although being based mainly on natural constituents too, since they undergo design, manipulation, refining, maturation and beneficiation in an artificial environment are hereafter called *designed and engineered peloids*.

Natural peloids are being less and less used in *Thermal spas* or *Health resorts* of all countries where traditionally have been used, and the reasons for that will be herewith referred to. It will be shown that the *designed and engineered peloids* could advantageously replace the use of *natural peloids* in *Thermal spas*, the advantages being threefold:

1. Simple composition based on raw materials, such as the almost monomineralic commercial clays *kaolin* or *bentonite* of both therapeutic and cosmetic grades, and the *natural mineral water* from a particular *Thermal spa* also recognized by their medicinal properties;

2. Easy incorporation of pharmacologically and medically recognized *functional active principles*, natural or synthetic, and inorganic or organic, into the matured paste prepared with *kaolin/natural mineral water* or *bentonite/natural mineral water*;

3. Simple and effective sanitary control.

Therefore, the identification, the control and the assessment of the *healing action* particularly provided by both *natural mineral water* and *functional active principles* existing in a certain *peloid* would be much easier with the simple composition of a *designed and engineered peloid* than with the complex composition of a *natural peloid*.

Key words: natural peloid, designed and engineered peloid, comparative advantages, peloid properties and functions, peloid functional additives

Peloides naturales *versus* peloides diseñados y fabricados

Resumen

El presente estudio pretende mostrar lo que diferencia a los *peloides naturales* de los *peloides artificiales*. Los últimos, aunque se basan principalmente también de componentes naturales, ya que se someten a diseño, manipulación, depuración y maduración, se elaboran

en un ambiente artificial, en adelante son denominados aquí *peloides diseñados y fabricados*.

Los *peloides naturales* están siendo cada vez menos empleados en los *balnearios o centros termales* de todos los países en los que tradicionalmente se han utilizado, las razones serán referidas. Se verá que los *peloides diseñados y fabricados* podrían reemplazar ventajosamente el uso de los *peloides naturales* en *centros termales*, siendo triples las ventajas:

1. Composición simple basada en materias primas, principalmente arcillas minerales comerciales *caolín* o *bentonita*, ambas con calidades terapéuticas y cosméticas, y el *agua mineromedicinal* de un centro termal concreto también reconocida por sus propiedades medicinales;

2. Fácil incorporación de *principios activos funcionales* reconocidos farmacológica y médicamente, naturales o sintéticos, y orgánicos o inorgánicos, en la pasta madurada preparada con *caolín/agua mineromedicinal* o *bentonita/agua mineromedicinal*;

3. Control sanitario simple y eficaz.

Por lo tanto, la identificación, el control y la evaluación de la *acción curativa*, sobre todo proporcionado, tanto por el *agua mineromedicinal* como por los *principios activos funcionales* existentes en un determinado *peloides* sería mucho más fácil con la simple composición de un *peloides diseñado y fabricados* que con la compleja composición de un *peloides natural*.

Palabras clave: peloides natural, peloides diseñado y fabricados, ventajas comparativas, peloides propiedades y funciones, peloides aditivos funcionales

REFERENCIA NORMALIZADA

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INTRODUCTION

Clay and mineral water are natural mineral resources which are considered as being major constituents of most *natural peloids* and *artificial peloids*. *Clay and mineral water* typologies are main conditioning factors of *peloid* properties and functions (Veniale, 1999; Tateo *et al.*, 2006; Gomes & Silva, 2007; Fraioli *et al.*, 2011; Gomes, 2013; Casás *et al.*, 2013; Gomes & Rautureau, 2013; Pozo *et al.*, 2013).

On the one hand, the use by humans of *clay* for healing and wellness purposes is most probably as old as mankind (De Vos, 2010; Gomes, 2013).

For medicinal purposes *clays* or *clay minerals* (the *clay* essential constituents) can be used internally or orally (e.g., the so-called *edible clays*) and externally or topically (e.g., the so-called *peloids*). Some types of *clay* are being used for therapeutic purposes, for instance as gastrointestinal ailments, as well as for nutritional and/or religious purposes (Bech, 1987, 1996; Robertson, 1996; Carretero, 2002;

Reinbacher, 1999, 2003; Gomes & Silva, 2007; Carretero *et al.*, 2006; Carretero & Pozo, 2007; Carretero & Pozo, 2009; Gomes *et al.*, 2008; Ferrel, 2008; Kikouama *et al.*, 2010). Also some types of *clay*, as is the case of the so-called *green clays*, have been the object of studies that demonstrate their *bactericide* character (Williams *et al.*, 2004; Williams *et al.*, 2008; Williams *et al.*, 2010; Williams *et al.*, 2011; Otto & Haydel, 2013).

Clays such as *kaolin*, *bentonite*, *palygorskite*, *sepiolite* and *talc* are reported in Pharmacopoeias, and being considered *medicines* they could participate in pharmaceutical formulations as *active principles* and/or *excipients* (Viseras & Lopez-Galindo, 1999; Lopez-Galindo & Viseras, 2004; Lopez-Galindo *et al.*, 2007; Viseras *et al.*, 2007; Viseras *et al.*, 2010). In particular, the use of *kaolin* and *bentonite* in human health, have been the object of significant research (Novelli, 1996, 1998, 2000; Cara *et al.*, 2000; Viseras *et al.*, 2006; Williams *et al.*, 2009; Casás *et al.*, 2011; Carretero *et al.*, 2013; Williams *et al.*, 2014).

Kaolin and *bentonite* in order to be used as pharmaceutical and cosmetic raw materials must be chemically, microbiologically and mineralogically innocent, what means that no human toxic and carcinogenic elements (As, Pb, Cd, Hg,...) or compounds (crystalline silica), and no pathogenic microorganisms (*Escherichia coli*, *Pseudomonas aeruginosa*, *Salmonella spp.*, and *Staphylococcus aureus*) should be present in the *clay*, unless in contents that fulfill the guidelines set forth by different Pharmacopoeias. For instance, according to US Pharmacopoeia (2007) and European Pharmacopoeia (2005), Pb and As contents in *bentonite* should not be higher than 40ppm and 5ppm, respectively. Also, according to Viseras *et al.*, 2006, the content of crystalline silica in *bentonite* must not be higher than 2%. Microbiologically *bentonite* should not be contaminated with *E. coli* and the total viable aerobic count must not be greater than 10^3 microorganisms per gram (US Pharmacopoeia, 2007, and European Pharmacopoeia, 2005).

Kaolin and *bentonite* are important *commercial special clays* showing distinct activities in terms of human health interaction, since their fundamental clay minerals, *kaolinite* and *montmorillonite*, respectively, significantly differ on structural, textural, physical and chemical properties. In comparative terms, *montmorillonite* is much more active, being characterized, for instance, for higher electric charge, higher specific surface area, higher ion exchange capacity and higher adsorption capacity.

On the other hand the *natural mineral water*, used for both internal and external applications in *Thermal spas*, is considered a *medicine* too.

Several diseases have been treated through the administration of *natural mineral water*, the health benefits mainly depend upon water chemical properties.

Maraver (2008) and Arribas (2008) specify the benefits for human health of the chemical characteristics of *natural mineral water* that they call *mineral medicinal water*, classified into six categories: **1.** Chlorinated (Cl^{-1} is the major anion); **2.** Sulphated (SO_4^{-2} is the major anion); **3.** Bicarbonated (CO_3H^{-1} is the major anion);

4. Gasocarbonic (with more than 250mg/L of free CO₂); 5. Sulfurous or “sulfúrea” (in Portuguese) (SH⁻¹ and S⁻² are the major anions); 6. Ferruginous (with more of 5mg/L of total Fe).

In Portugal most *natural mineral waters* used in *Thermal spas* are of sulfurous or “sulfúrea” (in Portuguese) typology, mainly occurring in the central region of the country.

Teixeira (2009; 2013b) enhances the physical and physical-chemical properties of Portuguese *natural mineral waters*, as well as the methodologies used for their application in “Crenotherapy” or Thermalism.

BASIC CONCEPTS

The official definition of *peloid* is still the one proposed by the *Société Internationale d'Hydrologie Médicale*, and adopted during the *IV^e Conférence Scientifique Internationale*, held in Dax, France, in 1949: “*peloids are natural products composed of a mixture of mineral water (sea water, salt lake water, and natural mineral water included), with organic or organic matter, resulting from geologic or biologic processes, or from both geologic and biologic processes, which are utilized for therapeutic purposes under the form of packs or baths*”. At the same meeting was adopted the *Classification Hydrologique Internationale de Pélóïdes* based on *peloids* origin, mineral water chemical nature and temperature, and maturation conditions.

Recently Gomes *et al.*, (2013) have proposed a classification of *peloids* into two major groups (Figure 1 and Figure 2) based on origin and maturation: **1.** *Natural peloids* defined “*as matured in situ healing mud or muddy dispersion composed of a complex mixture of fine-grained materials, mineral water or sea water, and often organic compounds from biological metabolic activity*”; **2.** *Artificial peloids* or *peloids s.s. (strictu sensu)* herein also called *designed and engineered peloids* because it could undergo more or less complex manipulation involving refining, maturation, formulation and beneficiation, defined “*as a healing mud or muddy dispersion resulting from the mixture clay/mineral water (natural mineral water or sea water) dressed and matured in an artificial environment, more precisely in open or closed tanks with or without stirring*”.

To the therapeutic use of *natural peloids* and of *artificial peloids* the same authors proposed the designations *mud therapy or mudtherapy* and *peloid therapy or pelotherapy*, respectively.

On the one hand, the *natural peloids* are being less and less used worldwide in *Thermal spas*, due to three principal characteristics and factors: **1.** Complex mineralogical and physicochemical composition of *peloid* solid, liquid and gas phases, making difficult to determine qualitative and quantitatively their constituents, as well as the identification of the *functional active principles*; **2.** Complex composi-

tion of the *microbiota* component, frequently bearing pathogenic microorganisms explained by *peloid* occurrence in natural open systems, hence easily contaminated; therefore the *peloid* requires a thorough and cautious sanitary safety control; **3.** The increasingly difficult access to *natural muds* (volcanic, marine, fluvial,...) because, in general, the mud deposits are located in environmentally protected areas.

Figure 1 – Peloids basic concepts (in Gomes, C. et al., 2013)

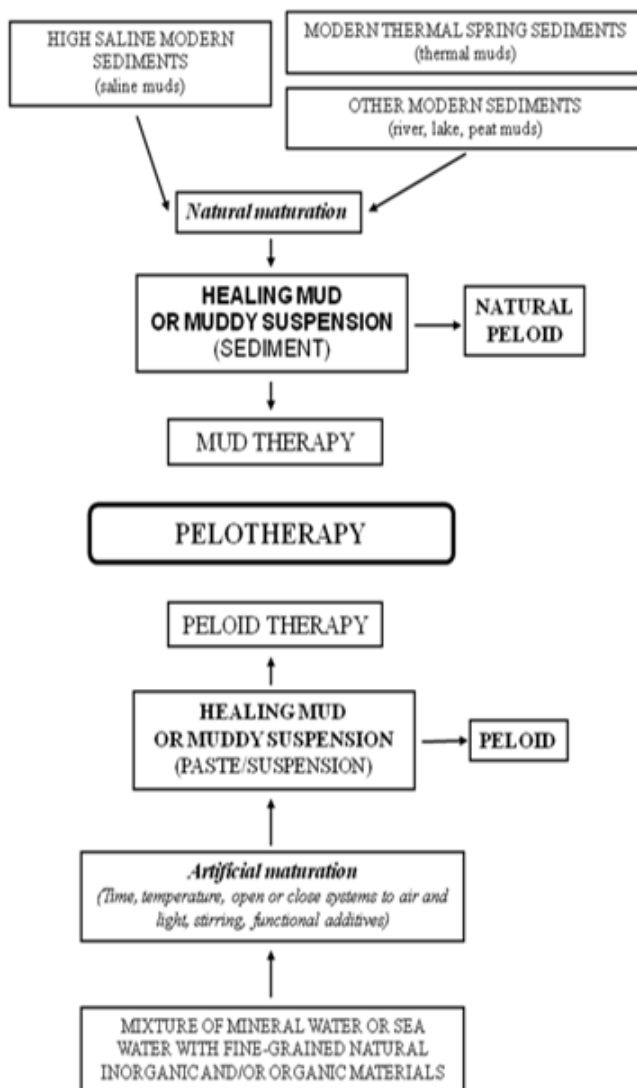
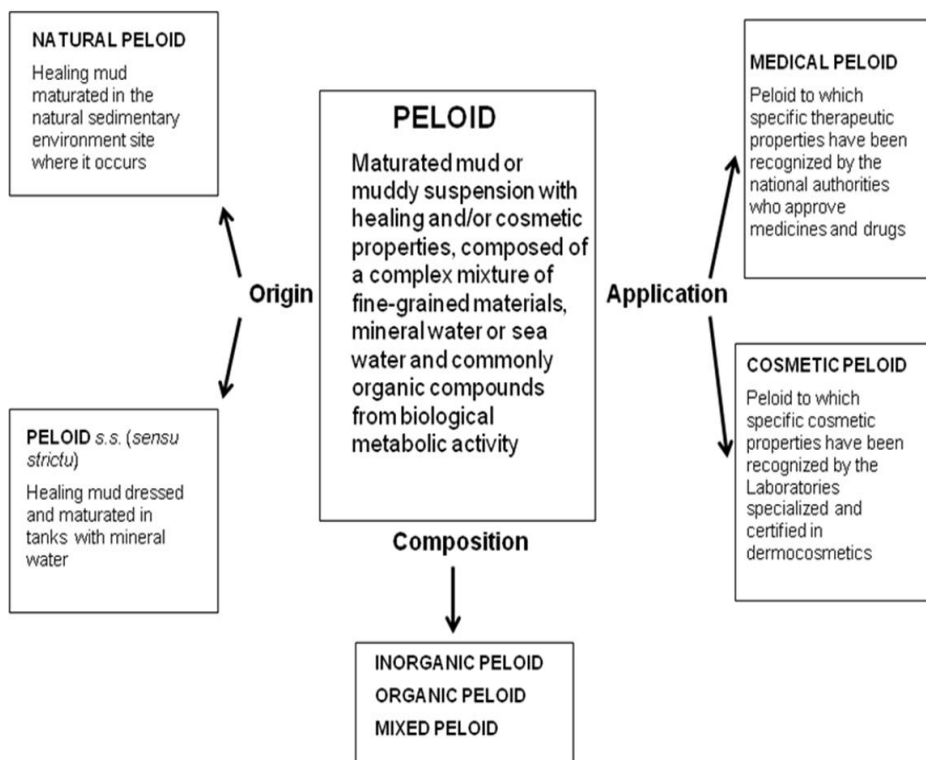


Figure 2 – Peloids origin, composition and application (in Gomes, C. *et al.*, 2013)

Depending upon composition (relative content of inorganic and organic constituents) both *natural peloids* and *artificial peloids* could be classified into three main groups: *essentially inorganic*; *essentially organic*; and *mixed inorganic/organic*.

Examples of *essentially inorganic peloids* are the popular mud packs and baths taken in the Bahrolmeyet Lake, in Palestina, the number one in the world in terms of salt concentration. The same happens in the Urumieh Lake, in Iran, the second lake in the world, in terms of salinity. Other example of *essentially inorganic peloid* used under medical counseling and supervision as mud-packs or mud baths, is the *volcanic mud* or *thermal mud* that occurs in the pool of Chanco, in Copahue (Argentina), the sulfur bearing warm mud (30-40°C) being deposited on the pool bottom and with origin in thermal springs (Ubogui *et al.*, 1991, 1998; Armijo *et al.*, 2008; Monasterio, 2008, 2009). Another example of *essentially inorganic peloid* is the so-called "lodo del Mar Menor", near to Murcia, in Spain, locally applied, either freely as mud baths, or as mud packs and mud baths in Thalassotherapy Centers.

Peat is a *mixed inorganic/organic peloid* in which the solid component is composed of an inorganic or mineral part that may reach 40%, and of an organic part very abundant in decaying plants. *Peat* or *turf* should be sterilized before application in the form of *peloid*. *Peat peloids* are widely used in northern Germany, Austria (particularly famous is the Neydharting peat, exported to many countries to be used for rheumatologic treatments and traumatismos related to sport activities), Belgium, the Czech Republic, Finland and in other eastern European countries.

The liquid component of *peat* or *turf* could be mostly lake water (chlorinated, or sulfidated, or ferruginous, or of other chemical nature), and sometimes sea water or artificially mineralized freshwater.

Biofilm or *bioglea* is an *essentially organic peloid* called *muffe* in Italy, and called *barégine* in France (due to first having been described in the thermal baths of Barèges), which is composed of algae and bacteria, and of a liquid component, generally reduced sulfur-bearing mineral water.

Also with regard to their applications, both *natural peloids* and *artificial peloids* could be classified into two major groups: *therapeutic* or *medical peloids* and *cosmetic peloids* (Figure 2).

On the other hand, the *designed and engineered peloids*, which really are *artificial peloids* or *peloids s.s (strictu sensu)*, since maturation is carried out in an artificial environment, for instance inside the *spas*, could have very simple composition based on the mixture of one thoroughly analyzed (mineralogically, physicochemically and microbiologically) almost *monomineralic commercial special clay (kaolin or bentonite)* of pharmaceutical or cosmetic grade, hence considered a *medicine*, and of one thoroughly analyzed (chemically and microbiologically) *specific natural mineral water* recognized by medical hydrologists by its healing properties for internal and/or external applications, hence considered a *medicine* too. In such simple mixture *additives*, both natural or synthetic, and inorganic or organic, pharmacologically and medically recognized as *medicines*, could be then incorporated.

Therefore, the *active principles* existing into the *designed and engineered peloid* could be more easily identified, and their functions and benefits could be better assessed.

Both *natural peloids* and *peloids s.s.* require to be empirically or scientifically recognized by their properties, good for both therapeutic and cosmetic applications.

Finally the topical use of *peloids* for therapeutic or cosmetic purposes is called *pelotherapy* (Veniale, 1999; Veniale *et al.*, 2004).

Presently no *natural peloids* or *artificial peloids* are used in Portuguese *Thermal spas*. Only “*peloids*” of the so-called *extemporaneous peloids* type are used in the *Thermal spas* of Alcafache and Caldas de Aregos.

However, *natural peloids* occurring in certain natural sites of the littoral Atlantic (Praia da Consolação, Praia da Parede, Praia do Meco, Praia de Porto de Mós) are still empirically used, and in the past *artificially matured natural peloids* have

been used in several *Thermal spas* (Caldas das Taipas, Termas de Vale de Cucos, Termas das Furnas).

Teixeira (2010, 2013a) presents a historical synthesis of the use of *peloids* in Portugal up to the last decade of the XX century when the use was discontinued, suggesting sanitary reasons to explain that discontinuity.

Then in the preparation of the *artificial peloid, natural mineral water*, as a rule *sulfurous water* or “*água sulfúrea*” (in Portuguese), of a specific *Thermal spa*, has been used. Naturally the *peloids* that incorporate this type of water will bear sulfur rich compounds recognized for their healing properties in case of respiratory, rheumatic and dermatologic diseases (Torresani, 1990; Costantino (2006); Baschini *et al.*, 2010).

STUDY GOALS

To show how complex is the composition of *natural peloids* which are multiphase and multi-component systems.

To show that, as a rule, when one *natural peloid* is used for a particular therapeutic purpose it is difficult to identify, either the *functional active principles* it contains, or to understand the *health benefits* it can provide.

To show that it is feasible to develop *designed and engineered peloids* characterized by simple composition based on a *maturated mixture* of an almost *monomineralic commercial special clay*, such as *kaolin* or *bentonite* both reported in Pharmacopoeias, and of a *specific mineral medicinal water*, into which could be incorporated, either natural pharmaceutical phytoproducts (e.g., liquid extracts or dry powders of *Equisetum arvense*, *Harpagophytum procumbens*, and *Filipendula ulmaria*, three medicinal plants reckoned by their *healing properties* in *rheumatic affections*), as well as liquid extracts or lyophilized powders of plants (e.g. *Arctium lappa*), and algae species (e.g. *Fucus spiralis*, *Fucus vesiculosus*, *Laminaria spp*, and *Ulva lactuca*) reckoned by their healing properties in skin affections and skin-care), or synthetic pharmaceutical products (e.g., *Voltaren* = Diclofenac sodic, liquid in ampoules) reckoned by their *healing properties* in *rheumatic affections* or by their beneficial effects in skincare.

FUNDAMENTALS OF NATURAL PELOIDS

Natural peloids are 3-phase systems: one solid phase (inorganic, organic or mixed inorganic/organic); one liquid phase (spring, river, lake or sea water); and one gas phase (H₂S, CO₂, NH₄, CH₄, Rn).

The inorganic component is mainly composed of a fine grained sediment called *clay*, itself a very complex system that consists of *clay minerals* and *non-clay minerals*.

The organic component is still mysterious, since comparatively to its inorganic counterpart it has been rather deficiently studied so far.

The liquid phase can also exhibit diverse and complex composition. The same happens with the gas phase.

Peloid Properties

Mean particle size, particle size distribution and particle aggregation are reflected on *natural peloid* or *peloid s.s.(strictu sensu)* thermophysical and rheological properties, such as: heat capacity, specific heat, heat diffusiveness, cooling rate, plasticity, viscosity, adhesivity, spreadability, impermeability, abrasivity.

Clay minerals nature and proportion, solid/liquid ratio, liquid nature and associated organic matter are reflected on *natural peloid* or *peloid s.s.* thermal properties, such as: specific heat, heat conductivity and cooling rate.

Chemical composition of solid, liquid and gas phases and total ion (cation and anion) exchange capacity and exchangeable ions are reflected on *natural peloid* or *peloid s.s.* chemical properties, such as: ion and molecule adsorption on a reversible way, and molecule absorption.

Chemical properties will be reflected on the transfer of chemical elements and compounds between *natural mineral water / clay minerals / organic matter* of both *natural peloid* or *peloid s.s.*, that is either natural sediment or artificial mixture, during maturation, as well as between the *peloid* and the human body along its application.

Microbiological content, and *pathogenic micro-organisms* speciation, are very important properties of *peloids*, either *natural peloids* or *peloids s.s.(strictu sensu)*.

As much complex is a *healing mud* or *peloid* in terms of composition, more time is required to accomplish its maturation and stability, and more attention and care requires its sanitary control and safety.

Peloid maturation is an important but complex process involving various physical and chemical parameters, and can provide physical, chemical, physicochemical and biological changes having consequences on *peloid healing activity*.

Peloid Functions and Benefits

Peloids are used under *medical counseling and accompanying* in *Thermal spas* and other *Health Resort Medicine Centers*, and their therapeutic effects are being particularly recognized in the fields of rheumatology and dermatology (Elkayam *et al.*, 2000; Bellometti *et al.*, 2005; Evcik *et al.*, 2007; Fioravanti *et al.*, 2007; Fioravanti *et al.*, 2010; Forestier *et al.*, 2010).

Peloid health benefits are being attributed to a combination of effects, and within these, the mechanical, the thermal and the chemical effects are currently emphasized.

Fioravanti *et al.* (2011) show and discuss the scientific evidences of the mechanisms of action on rheumatic diseases of the use of *thermal natural mineral water and peloids*.

Clinical evidence based on epidemiological studies, and available on specialized literature, emphasizes the benefits, at least short-term benefits, of *mudtherapy* and *pelotherapy* on the degenerative *rheumatism* or *osteoarthritis*, the *inflammatory rheumatism*, the *fibromyalgia*, the *spondylitis* and the *chronic back pain*, within other rheumatic affections, taking advantage of *peloids* analgesic, anti-inflammatory, anti-oxidant and antimicrobial action (Elkayam *et al.*, 1991; Bellometti *et al.*, 2005, 2007; Ceccarelli *et al.*, 2010; Codish *et al.*, 2005; Britschka *et al.*, 2007; Giannitti *et al.*, 2008; Constant *et al.*, 1995; Strauss-Blasche *et al.*, 2002).

Also, there is evidence of the positive effects of *peloids* on dermatological affections, especially on *psoriasis* (Costantino *et al.*, 2005, Clijsen *et al.*, 2008) and acne and seborrhoea, as well as on skin care functions (cleansing, degreasing, exfoliating, hydrating, tonifying, and reaffirming) (Portugal-Cohen *et al.*, 2009; Carbajo *et al.*, 2010).

Barros *et al.* (2010) and Pena Ferreira *et al.* (2010a, 2010b) have shown the effectiveness of several formulations involving *bentonite* from Porto Santo island, Madeira archipelago, either with cleansing and clarifying effects in facial masks, or with anti-cellulite effects.

Also, Meijide *et al.* (2010) and Arribas *et al.* (2010a, 2010b) have studied and assessed respectively the effects on the mechanical and functional characteristics of skin of the application of *extemporaneous peloids* based on *bentonite* mixed with both sulfurous mineral medicinal water and *sea water*, and the clinical effects (expressed by scaling off and inflammation) on *psoriasis* of the *thermal natural peloid* from La Toja (Spain).

Maraver (2013) discloses a very interesting and comprehensive review about the research being carried out in recent years on the relevant mechanisms of action (thermal, biologic and immunological) involved in *pelotherapy*.

As a rule, clinical evidences do not identify the *active principles* responsible for therapeutic and dermatologic benefits, certainly due to the complex composition of both *natural peloids* and *artificial peloids* being used.

The inorganic component of one *peloid* could be constituted by various clay minerals and non-clay minerals and the organic component could be constituted of up to few dozens of organic compounds (Suárez *et al.*, 2011; Gomes *et al.*, 2013a; Gomes *et al.*, 2013b). Such complex composition could be demonstrated in the two examples of *natural peloids* described below.

Examples of complex *natural peloids*

1.- Characteristics of the volcanic mud from Balneário da Coroa, S. Miguel island, the Azores (Figure 3, *in*: Gomes *et al.*, 2013b).

Figure 3 – Tank containing the volcanic mud used in Balneário da Coroa, São Miguel island



Solid phase:

Inorganic constituents: Clay minerals (kaolinite, illite, smectite), non-clay minerals (cryptocrystalline silica, feldspars, alunite, S).

Organic constituents: Organic matter (~1.5% in weight) consists of several polar and non-polar compounds: long chain alkanes (C15-C29), long chain alkenes, esters, aldehydes, and biofilms.

Microbiota: includes several pathogenic microorganisms.

Liquid phase:

Hydrothermal water: 95°C, pH=3, Cl⁻¹, F⁻¹, CO₃⁻², HCO₃⁻¹, SH⁻¹, SO₄⁻², Na⁺¹, K⁺¹, Ca⁺², Mg⁺², Sr⁺², and several trace elements.

Gas phase:

SO₂, CO₂, CH₄, Rn.

2.- Characteristics of the salt pans saline mud from Santiago da Fonte, Aveiro, Portugal (Figure 4, *in*: Gomes *et al.*, 2013a).

Figure 4 – Salt pans and saline mud from Santiago da Fonte, Aveiro



Solid phase:

Inorganic constituents: Clay minerals (illite, smectite, interstratified illite-smectite, kaolinite, chlorite), *non-clay minerals* (quartz, feldspars, micas, calcite) and sulfur compounds.

Organic constituents: Organic matter (~4.5% in weight) consists of esters that might derive from fatty acids, long chain alkenes, cycloalkenes, and long chain alkanes and ketones being originated from plant degradation or being by-products from microbiota action.

Microbiota: includes several pathogenic microorganisms.

Liquid phase:

High saline sea water (density: 1.27).

Gas phase:

H₂S, CO₂, CH₄, NH₄

PELOIDS S.S. (STRICTU SENSU) OR DESIGNED AND ENGINEERED PELOIDES

Natural peloids, in general, as was said before are complex mixtures of solid, liquid and gas phases, each one containing diverse and specific components. The complexity referred to makes difficult not only the identification of the *healing active principles*, but also the understanding of the healing process, and the control of both composition and sanitary state.

Also the access to *natural peloids* is becoming more and more difficult due to environmental restrictions (the deposits are, as a rule, located in environmental protected areas).

Peloids s.s. or *designed and engineered peloids* could be produced, either inside a specialized manufacture unit, or inside the spa facilities where they are applied.

The mixture consisting of an almost *monomineralic commercial special clay* (e.g., *kaolin* or *bentonite*)/*specific mineral medicinal water* undergoes maturation, manipulation, and beneficiation with the incorporation of *healing functional active additives* (analgesic, anti-inflammatory, anti-oxidation, anti-cellulite, anti-psoriasis, anti-acne,...), either natural or synthetic, inorganic or organic. It is convenient to emphasize that the initial mixture *clay/natural mineral water* do not contains organic compounds, fact that could enable the easier identification of the *active functional principles*.

Specific natural salts rich in bioessential chemical elements, for instance hydrous magnesium sulfates (*epsomite* and *hexahydrate*) derived from the evaporation of the sea water collected in salt pans, could be added as *functional additives* to the mixture.

Also, organic compounds, such as some long-chain alkanes, long-chain esters and steroids being recognized by analgesic, anti-oxidant, anti-inflammatory and antineoplastic properties (Curri *et al.*, 1997; Odabasi *et al.*, 2007; Tserenpil *et al.*, 2010; Suárez *et al.*, 2011; Cavaleiro, 2013) could be used as *functional additives*.

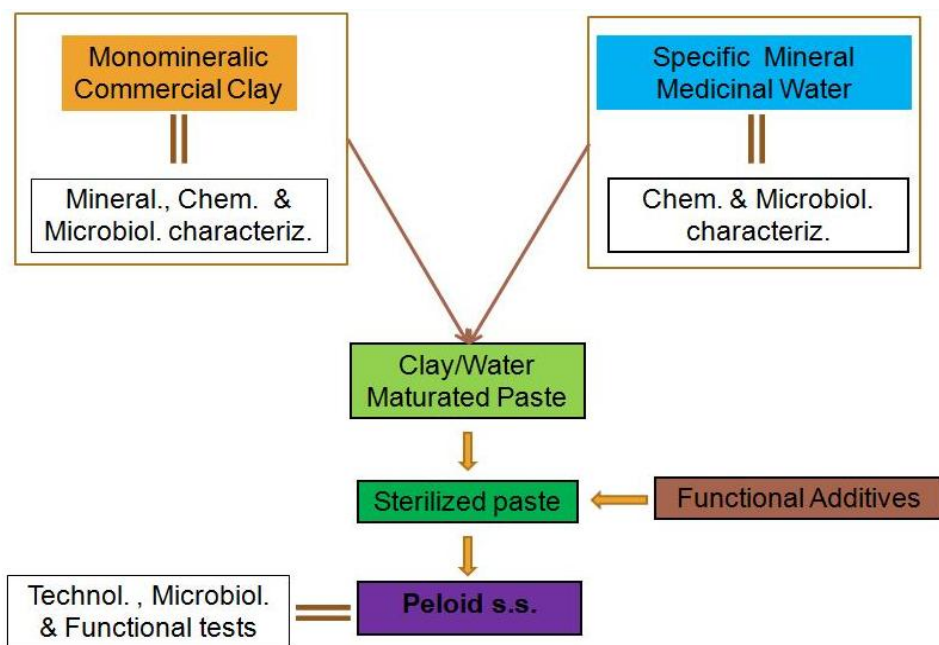
Also, organic compounds, such as some long-chain alkanes, long-chain esters and steroids being recognized by analgesic, anti-oxidant, anti-inflammatory and antineoplastic properties (Curri *et al.*, 1997; Odabasi *et al.*, 2007; Tserenpil *et al.*, 2010; Suárez *et al.*, 2011; Cavaleiro, 2013) could be used as *functional additives*.

Figure 5 shows the processing flow sheet that includes the preparation stages of the *designed and engineered peloid*, involving fundamental analytical studies of both solid and liquid phases, and of the matured mixture too. However, as a rule, the suppliers of both commercial therapeutic and cosmetic grades of *kaolin* and *bentonite* provide the corresponding relevant analytical data. Also, as a rule, the analytical data of the selected *natural mineral water* it is known and available too.

To achieve the equilibrium in the mixture *kaolin* or *bentonite/natural mineral water* 2-3 weeks of maturation time are sufficient. Such equilibrium will be attained and identified when no significant changes are found in the results of microchemi-

cal analyses carried out, along maturation, in the liquid phase resulting from the interaction *natural mineral water/kaolin* or *bentonite*. When the solid phase is *kaolin* the maturation time is significantly less than when the solid phase is *bentonite*.

Figure 5 – Flow sheet showing the preparation phases of *designed and engineered peloids*



Depending on its nature, natural or synthetic, the *healing functional additives* could be incorporated during the paste maturation or just before its application.

Carretero (2013) has proposed and recommended a methodology to be used in the preparation of *artificial peloids* of the type we have classified as *designed and engineered peloids*, starting with the mineralogical, chemical, physical and microbiological analyses of the solid phase (clay) and with the chemical, physicochemical and microbiological analyses of the liquid phase (mineral medicinal water). After the blending of *clay* with the *mineral medicinal water* the author enhances the importance of the analysis of the peloid interstitial liquid phase.

Fernandez-González *et al.* (2013) following the study of one *artificial peloid* prepared with a blend of two clays (*kaolin* and *bentonite*) and one *mineral medicinal water* found that 1 month of maturation time would be sufficient to provide the *optimum peloid*.

The application of *peloids s.s.* always requires medical counseling and supervision. Also, periodical sanitary control (identification and quantification of pathogenic microorganisms) is always required.

CONCLUSIONS

Natural peloids, in general, are complex mixtures of solid, liquid and gas phases, each one containing diverse and specific components. The complexity referred to makes difficult not only the identification of the *healing active principles*, but also the understanding of the *healing process* and the control of both composition and sanitary state. Also the access to *natural peloids* is becoming more and more difficult due to environmental restrictions (the deposits are, as a rule, located in *environmental protected areas*).

Natural peloids, in general, are complex mixtures of solid, liquid and gas phases, each one containing diverse and specific components. The complexity referred to makes difficult not only the identification of the *healing active principles*, but also the understanding of the *healing process* and the control of both composition and sanitary state. Also the access to *natural peloids* is becoming more and more difficult due to environmental restrictions (the deposits are, as a rule, located in *environmental protected areas*).

Thermal spas could, either order and buy, or even design and develop genuine, specific and differentiated *peloids s.s.* for specific therapeutic and skincare uses. The *manipulated peloids s.s.* after getting their *healing properties* assessed and validated by *medical hydrologists*, could deserve the due approval as *new medicines* from the competent Institutions.

If certain *natural mineral waters* used in *Thermal spas* for external and internal applications are being considered *medicines* by *medical hydrologists*; if certain *clays*, such as *kaolin* and *bentonite* reported in Pharmacopoeias as *active principles* and *excipients* are being considered *medicines*; if certain natural and synthetic chemicals, pharmacologically and medically recognized by their specific and functional health benefits are considered *medicines* too, even after their incorporation into the clay/water paste; naturally, the *peloids s.s.* or *designed and engineered peloids* which are based on controlled mixtures of the referred to materials should be considered *medicines* too.

The simple composition of these *peloids* requires short-term maturation, and the maturated paste constituted of the mixture *specific clay/specific natural mineral water* could be kept in sanitary safe conditions.

Whenever required for the topical treatment of one specific health affection, the medically indicated *active and specific functional additive* could be added to a well dimensioned sample of that mixture.

The *peloid activity* will depend more on the chemistry of both the *specific natural mineral water* and the *specific functional additives* than on the physical, physicochemical and chemical characteristics of the *clay*.

Clay will act mainly as an heat conservation and transfer agent, and as a delivery vehicle that facilitates the controlled liberation and the lasting action of part of the *active healing principles* (specific chemical elements and compounds existing both into the *water* and into the *additives*) which became fixed on a reversible way at the clay particle surfaces during the peloid preparation.

In the authors opinion the future of *peloids* and *pelotherapy* would depend much on the manipulation and use of *designed and engineered peloids* that besides being applied in *Thermal spas* could well be also used in *Thalassotherapy Centres*, where *sea water* would take the place of *natural mineral water*.

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